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# Synchronization of Coupled Oscillators on a Two-Dimensional Plane

Dameng Guo,<sup>[a]</sup> Yong Qing Fu,<sup>[b]</sup> and Bo Zheng<sup>\*[a]</sup>

**Abstract:** The effect of the transfer rate of signal molecules on coupled chemical oscillators arranged on a two-dimensional plane was systematically investigated in this paper. A microreactor equipped with a surface acoustic wave (SAW) mixer was applied to adjust the transfer rate of the signal molecules in the microreactor. The SAW mixer with the adjustable input powers provided a simple means to generate different mixing rates in the microreactor. A robust synchronization of the oscillators was found at an input radio frequency (RF) power of 20 dBm, with which the chemical waves were initiated at a fixed site of the oscillator system. With the increasing input power, the frequency of the chemical waves was increased, which agreed well with the prediction given by the time-delayed phase oscillator model. Results from the finite element simulation agreed well with the experimental results.

## Introduction

In this work, we developed a microfluidic platform integrated with a surface acoustic wave (SAW) mixer to study the effect of the transfer rate of the signal molecules on the behaviors of oscillator population in a microreactor. In nature, bacteria and many other types of cells communicate via the transfer of signal molecules,<sup>[1]</sup> leading to collective behaviors such as quorum sensing<sup>[1a, 2]</sup> and biofilm formation.<sup>[3]</sup> The switching from the individual to the collective behaviors depends on the cell density<sup>[3a]</sup> and the interaction intensity between the cells, e.g. the transfer rate of the signal molecules.<sup>[4]</sup> This transfer rate plays a critical role on the collective behaviors in the network systems such as the propagation of electrochemical perturbations along axons<sup>[5]</sup> and cell-to-cell communication based on diffusion coupling of the signal molecules.<sup>[1c]</sup> In these network systems, certain time is needed for a signal molecule to travel from one oscillator to another oscillator, and the time is of the same order or larger than the typical time scales of the individual oscillator dynamics.<sup>[6]</sup> The signal transfer time dependent behavior of the network systems have a remarkable range of dynamics<sup>[7]</sup> but is generally difficult to study due to the complicated nature of the cells in the systems.

To study these network systems, the resin particles doped with the catalyst of the Belousov-Zhabotinsky (BZ) reaction have

been used as the model oscillators.<sup>[4, 8]</sup> The effect of the transfer rate of the signal molecules on the synchronization of the chemical oscillators was previously investigated by adjusting the stirring rate in the bulk reaction.<sup>[8b]</sup> However, applying a continuous-stirring system makes it difficult to observe the propagation of the signal between the oscillators. By contrast, an unstirred two-dimensional oscillator system facilitates *in situ* monitoring the individual oscillator and tracking the signal transfer.<sup>[8b, 8d, 9]</sup> Nevertheless, there are few techniques available which can enhance the transfer rate of the signal molecules with registered positions of the oscillators. As a result, the study of the behaviors of populations of coupled oscillators is limited.

The surface acoustic wave microfluidics provides a simple alternative way of manipulating and actuating fluids in microdevices,<sup>[10]</sup> which could enhance the transfer rate of molecules in oscillator systems.<sup>[11]</sup> The surface acoustic waves travel along the surface of the piezoelectric substrate with the typical wavelength in the 10-100  $\mu\text{m}$ ,<sup>[12]</sup> resulting in the acoustic streaming in the fluid without new chemical processes.<sup>[11]</sup> Using the SAW based microfluidic techniques,<sup>[13]</sup> the reactor could be specifically designed on a superstrate, under which the SAW chips could generate the acoustic radiations to the reactor.<sup>[12, 14]</sup> Herein, a microfluidic platform integrated with a SAW<sup>[10a, 10e, 10f]</sup> mixer was introduced to investigate the behaviors of the coupled oscillators arranged on a two-dimensional plane. The mixer can conveniently change the transfer rate of the chemical species in the microreactor, thus providing a useful means to study the behaviors of the oscillator population.

## Results and Discussion

### The detection platform

The cation-exchange particles (50 W  $\times$  4 250 - 300, Dowex) were sieved between 250 meshes and 280 meshes to achieve a narrow size distribution (Figure 1D). The particles were then doped with  $\text{Ru}^{2+}$  with a final concentration of  $1.5 \times 10^{-6} \text{ mol/g}$ <sup>[9b]</sup> to be used as the oscillators. The microreactor was a PDMS-glass hybrid device, made of a PDMS slab and a glass substrate (supporting information). The glass substrate helped reduce the accumulation of  $\text{Br}_2$  generated from the BZ reaction in the microreactor, as the  $\text{Br}_2$  can be absorbed by the PDMS and inhibits the BZ reaction.<sup>[9c, 15]</sup> The mixing device was fabricated in Teflon (Figure S1) to make the mixing of the BZ reagents and coupled with the microreactor through Teflon tubing (Figure 1A). The Teflon device had an excellent corrosion resistance to the BZ reactants and the reaction intermediates. The flow rate of the BZ mixture was set at 0.33  $\mu\text{L}/\text{min}$ . The hybrid microreactor was placed on the SAW platform. A chirped SAW device was purposely fabricated on a 128° Y-Cut  $\text{LiNbO}_3$  substrate, and has variable resonant frequencies from 20 MHz to 40 MHz for driving the liquid.

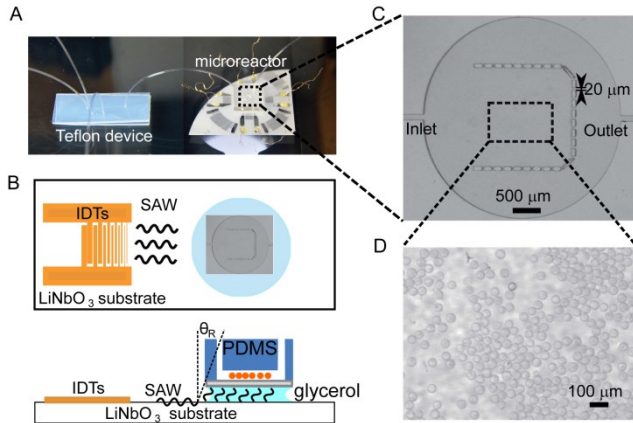
The input power on the IDTs was supplied from an RF signal generator (Agilent, E4421B) multiplied by a 43 dB amplifier

[a] Dr. D. Guo, Prof. B. Zheng  
Department of Chemistry  
The Chinese University of Hong Kong  
Shatin, New Territories, Hong Kong  
E-mail: [bozheng@cuhk.edu.hk](mailto:bozheng@cuhk.edu.hk)

[b] Dr. Richard Y. Q. Fu  
Department of Physics & Electrical Engineering, Faculty of  
Engineering and Environment  
Northumbria University  
Newcastle Upon Tyne, UK NE1 8ST.

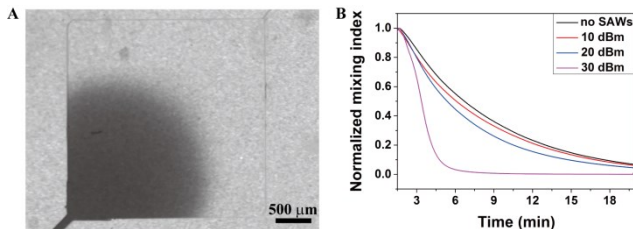
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(Mini-Circuits, LZY-22+). The input RF signal was fixed at 20 MHz. Glycerol was added between the LiNbO<sub>3</sub> substrate and the microreactor as the medium for the propagation of the acoustic radiations with the Rayleigh angle,  $\theta_R \approx 23^\circ$  (Figure 1B).<sup>[12]</sup> To decrease the photoeffect<sup>[15-16]</sup> on the BZ reaction, the illuminating light was adjusted to minimum intensity that allowed a clear photo to be taken using a CCD camera (1.4 MP monochrome, SPOT) at the exposure time of 150 ms.



**Figure 1.** A) Picture of the microfluidic platform. B) Scheme of the SAW mixer. The glycerol liquid was sandwiched between the PDMS device and the substrate. The orange spheres were the resin particles doped with Ru<sup>2+</sup>. C) Microscopic photo of the microreactor marked in dash line in A. D) Microscopic photo of the resin particles.

### The mixing in the presence of the SAWs



**Figure 2.** A) Image of red dye in the micro-reactor before mixing. B) Plot of normalized mixing index versus time.

The SAWs radiations leaked into the microreactor caused the chaotic advection, which enhanced the mixing of the chemical species, and thus altered the transfer rate of the chemical species.<sup>[11]</sup> The mixing in the microreactor could be indicated by the mixing of red food dye (McCormick) in the presence of the SAWs (Figure 2A). The red dye was injected into the microreactor at a rate of 0.5  $\mu\text{L}/\text{min}$  for 80 s to cover about half of the reactor. The input RF powers of 10, 20, and 30 dBm on the IDTs were then applied to induce SAW radiations into the microreactor.

The mixing index<sup>[11]</sup> to describe the mixing of the red dye in water could be determined from the pixel analysis on the photos

of the microreactor, which were taken by the CCD camera. The standard deviation  $\sigma$  of the pixel grayscale was obtained from the Matlab software:

$$\sigma = \sqrt{\left(\frac{1}{N}\right) \sum_{i=1}^N \left(\frac{P_i - \bar{P}}{\bar{P}}\right)^2}, \text{ for } 0 \leq P_i \leq 1 \quad (1)$$

where  $P_i$  is each pixel grayscale of the photo,  $\bar{P}$  is average grayscale of the pixels and  $N$  is the pixel number. The normalized mixing index (NMI), which was varied between one (before mixing) and zero (completely mixed), was defined as

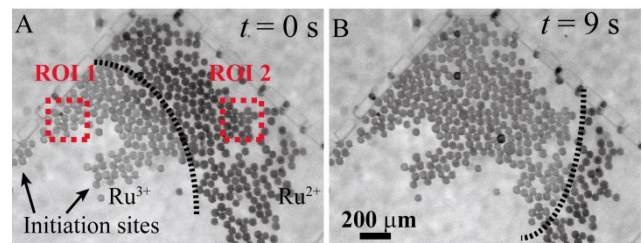
$$NMI = \frac{\sigma - \sigma_{final}}{\sigma_{initial} - \sigma_{final}}, \quad (2)$$

where  $\sigma_{initial}$  and  $\sigma_{final}$  are the standard deviation of the pixel grayscale before mixing and 30 min after mixing, respectively.

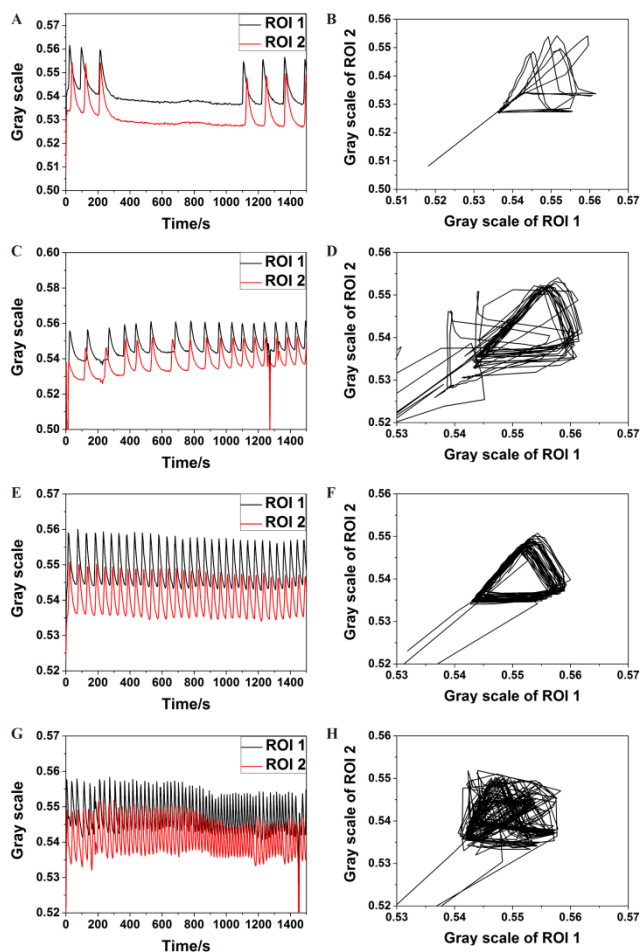
As shown in Figure 2B, in the presence of the SAWs with an input power of 30 dBm, the dye was observed to quickly disperse homogeneously throughout the reactor within 6 min, whereas the dye did not disperse homogeneously even after half an hour in the absence of the SAWs. The larger the input power, the faster the mixing was observed.

### The chemical wave propagation and frequency under different amplitudes of the SAWs

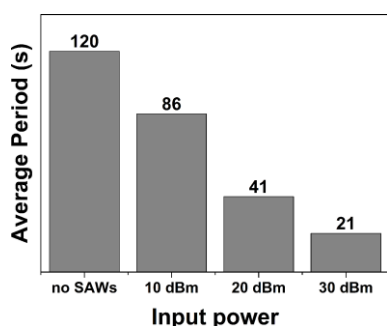
The RF input power on the IDTs was set at 10 dBm, 20 dBm and 30 dBm to obtain different transfer rates of the chemical species in the microreactor. After the resin particles were loaded, the BZ mixture was flowed into the microreactor for 30 min to obtain robust oscillations of the oscillators before the input power was turned on. In the two-dimensional oscillator system, the oscillators could present chemical waves<sup>[15]</sup> with appropriate conditions such as the recipe of the BZ mixture, the oscillator density in the BZ mixture and so on.<sup>[17]</sup> A robust chemical wave activity would emerge when the oscillators were oscillating at the same frequency with different phase, i.e., frequency synchronized (Figure 3, Movie S1).



**Figure 3.** Wave propagation through the oscillators. The dashed line separates the oscillators with Ru<sup>3+</sup> (left) and Ru<sup>2+</sup> (right)



**Figure 4.** The oscillation and the entrainment or the lack of the entrainment under RF powers of the SAWs: no SAWs (A and B), 10 dBm (C and D), 20 dBm (E and F) and 30 dBm (G and H). The plots in right column are phase-plane plots of the dynamical system. If the oscillators are frequency synchronized, i.e., the oscillators are oscillating with the same frequency but different phase, the trajectory will be a closed loop.



**Figure 5.** Periods of the oscillators under different input powers.

To investigate the chemical wave propagation, two regions of interest (ROI 1 and ROI 2, Figure 3A) were taken into account. Typically the chemical waves were initiated at certain oscillators, i.e., initiation sites, and then propagated to the surrounding

oscillators in all directions. If no SAWs were applied (Figure 4A and B, Movie S1), the transfer of the molecules in the microreactor depends only on the molecular diffusion. During the whole observation period, the chemical wave activity was unstable and even unobservable for a few minutes sometimes, and the initiation sites kept changing. When the input SAW power was increased to 10 dBm, the chemical waves were initiated at different sites of the oscillator system and then propagated to the surrounding oscillators. A robust wave activity occurred when the input SAW power was increased to 20 dBm (Figure 4E, Movie S1), and the chemical waves were initiated at the same site of the oscillator system and then propagated to the surrounding oscillators. The closed loop in the plot of ROI 1's grayscale against ROI 2's (Figure 4F, Movie S1) revealed that ROI 1 and ROI 2 were fully entrained.<sup>[18]</sup> When the input RF power was increased to 30 dBm, the entrainment between the ROI 1 and ROI 2 was broken (Figure 4G and H, Movie S1), and the wave initiation sites were again shifted to different sites. We also observed that with the increasing input power, the frequency of the chemical waves was increased (Figure 5).

The initiation site of the chemical waves was dependent on the local concentration of the activator  $\text{HBrO}_2$ <sup>[19]</sup> and the properties of oscillators.<sup>[20]</sup> In an inhomogeneous medium, the local accumulation of the activators could cause shifting initiation sites.<sup>[21]</sup> Therefore, without the SAW radiations, the chemical waves were initiated on different sites, and then propagated to the other oscillators. The disappearance of the chemical waves (Figure 4A, Movie S1) suggested the existence of local convections induced by the flow through the microreactor.<sup>[22]</sup>

When the input power was increased to 20 dBm, the molecules would be dispersed homogeneously more quickly throughout the reactor, so that the homogenized concentration of the activator,  $\text{HBrO}_2$ , surrounding the oscillators tended to decrease the heterogeneity of the oscillators. The initiation site was then dependent on the properties of oscillators, such as the size, concentration of the catalyst, the surrounding oscillators<sup>[20]</sup> and so on,<sup>[15, 19]</sup> which were consistent when the oscillator system was established. Therefore, the chemical wave activity was more robust when the input power was increased to 20 dBm, as the chemical waves were initiated at the same site of the oscillator system. However, if the input power was further increased to 30 dBm, the chemical wave activity became unstable, probably due to the disturbance by a strong acoustic force on the oscillators and surrounding environments of the oscillators.<sup>[10f]</sup>

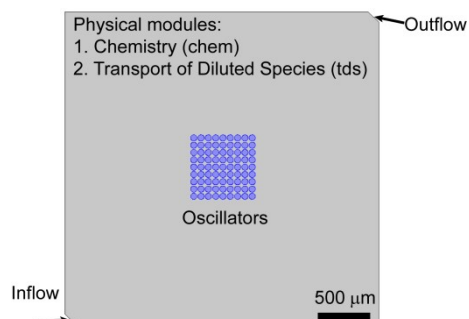
### Simulation by COMSOL

COMSOL software was employed to simulate the oscillator population in the experiment (supporting information). Instead of the chaotic advection, the turbulent mixing was introduced in the simulation to change the mass diffusivity of the molecules, i.e., transfer rate of the molecule, as the turbulent mixing module was already contained in the software and the mass diffusivity was the key point no matter what means was used to change the diffusivity. The turbulent mass diffusivity  $D_{T,i}$  of the molecule  $i$  is defined as

$$D_{T,i} = \frac{v_T}{Sc_T} \quad (8)$$



where  $\nu_T$  is the eddy viscosity and  $Sc_T$  is the turbulent Schmidt number. The turbulent Schmidt number of water, 340 at 300 K,<sup>[23]</sup> was chosen as the Schmidt number of the BZ solution in the microreactor to simplify the simulations. The transfer rate of the molecules can be adjusted by changing the eddy viscosities.

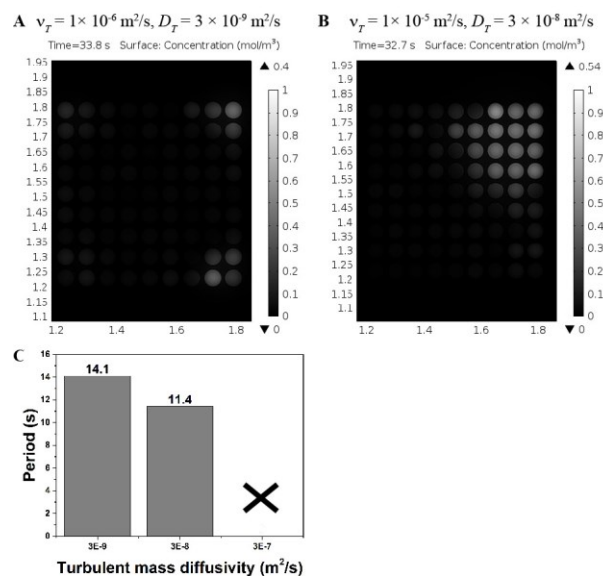


**Figure 6.** Image of 81 oscillators outputted from the COMSOL software.

As shown in Figure 6, 81 circles with a diameter of 60  $\mu\text{m}$  represented 81 oscillators in a two-dimensional 3 mm  $\times$  3 mm reactor. The number of the oscillators was limited by the computing power. Two physical modules, Chemistry (CHEM) and Transport of Diluted Species (TDS), were applied to simulate the reaction and mass transport of the molecules in the reactor, respectively. The heterogeneity of the oscillators was controlled by a normal random function with the mean value of 4 and a standard deviation of 1, which provides the concentration information of the immobilized catalyst inside the oscillators at mM scale. The chemical model of the BZ reaction was chosen from Försterling's work (supporting information).<sup>[24]</sup> A flow of the BZ mixture, the components of which were consistent with those in the experimental work, was introduced in the model to maintain the oscillations. The inflow boundary condition was set as concentration constraint, which means the concentrations of the BZ mixture were fixed at the inflow.

The value of  $\nu_T$  was initially set as  $10^{-6} \text{ m}^2/\text{s}$ , where the turbulent mass diffusivity was set as  $3 \times 10^{-9} \text{ m}^2/\text{s}$ . In the simulation results (Figure 7A, Movie S2), the chemical waves were initiated at the center of the oscillator array at the time of 20.0 s, but the chemical wave changed the initiation sites to the four corners of the oscillator array at the time of 33.8 s. The simulation agreed well with the experimental results that the initiation sites appeared at different sites of the array when the SAW input power was absent or 10 dBm.

The value of  $\nu_T$  was then increased to  $10^{-5} \text{ m}^2/\text{s}$ , where the turbulent mass diffusivity was set as  $3 \times 10^{-8} \text{ m}^2/\text{s}$  in the reactor. In the simulation results (Figure 7B, Movie S3), the chemical waves were initiated at the top-right corner of the array of the 81 oscillators and propagated at a period of 11.4 s, which agreed well with the experimental results that the initiation sites appeared at the same locations of the array when the input power was increased to 20 dBm.



**Figure 7.** A) Image of the simulation at time of 33.8 s and  $\nu_T$  of  $10^{-6} \text{ m}^2/\text{s}$ , which is the kinematic viscosity of water at 20 °C. B) Image of the simulation at time of 32.7 s and  $\nu_T$  of  $10^{-5} \text{ m}^2/\text{s}$ . C) Periods of the oscillators under different turbulent mass diffusivities.

The value of  $\nu_T$  was finally increased to  $10^{-4} \text{ m}^2/\text{s}$ , where the turbulent mass diffusivity was set as  $3 \times 10^{-7} \text{ m}^2/\text{s}$  in the reactor. In the simulation, the oscillators did not show oscillations in the simulation, as it is difficult to accumulate the activator,  $\text{HBrO}_2$ , at a high mass diffusivity in an open microreactor. However, in the experimental work, the disappearance of the oscillations did not occur due to the relatively low input power of the SAW mixer.

The period of the oscillators in the simulation was decreased from 14.1 s at  $\nu_T = 10^{-6} \text{ m}^2/\text{s}$  to 11.4 s at  $\nu_T = 10^{-5} \text{ m}^2/\text{s}$  (Figure 7C), which agreed well with the experimental results that the period of the oscillators was decreased with increasing transfer rate. The simulation showed that the chemical waves with SAW were initiated at the same site of the oscillator array at a proper high transfer rate while they were initiated at different sites of the oscillator array at a low transfer rate, and the frequency of the oscillators was increased with increasing the transfer rate of the molecules. These results agreed well with the experimental observations.

## Conclusions

The effect of the transfer rate of signal molecules on the BZ reaction oscillators on a two-dimensional plane was investigated by a microfluidic device integrated with a SAW mixer. It was convenient to induce different mixing rates into the microreactor by using the SAW mixer with adjustable input power of the IDTs. The results showed only a proper range of the transfer rate could support a robust synchronization of the time-delayed-interactions oscillators, which is common in the biologic systems, e.g., the high conductivity of the connexins guarantees a strong coupling strength to synchronize the population of heart muscle

cells.<sup>[25]</sup> On the other hand, the higher transfer rate was found to increase the frequency of the chemical waves and the susceptibility of the system to the perturbations in the reactor. In the neural network, enhanced activity of neuron population usually causes higher frequency or increased amplitude of the neural oscillation<sup>[26]</sup> and decreases the delay time, which then increases the susceptibility of the neural network.<sup>[27]</sup> Here a simple chemical model system was able to present similar communication dynamics, providing a simple approach to study the complicated networks in nature. The results on how the transfer rate affects the robustness of the synchronization highlighted the significance of the transfer-rate-related factors, such as convection and temperature in the behavior of the network systems. By using the microfluidic platform combined with theoretical analysis and numerical simulation, we expect to gain more insight on the signal-transfer related phenomena of network systems.

## Experimental Section

### Chemical and reagents

Tris(2,2'-bipyridyl)dichlororuthenium(II)hexahydrate ( $\text{Ru}(\text{bpy})_3\text{Cl}_2$ ) and sodium bromate ( $\text{NaBrO}_3$ ) (Aldrich), malonic acid (MA) (National Chemicals Import and Export), and nitric acid (63%, BDH) were used in as-received conditions. The BZ mixture contained 0.16 M malonic acid (MA), 0.5 M  $\text{NaBrO}_3$ , 0.77 M  $\text{H}_2\text{SO}_4$  and 0.06 M  $\text{NaBr}$ .

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**Keywords:** chemical wave activity • transfer rate • the Belousov-Zhabotinsky reaction • microfluidics • surface acoustic wave

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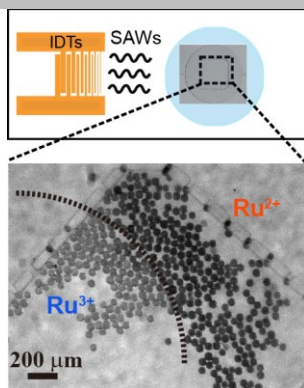
## Entry for the Table of Contents (Please choose one layout)

Layout 1:

### ARTICLE

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The effect of the transfer rate of signal molecules on coupled chemical oscillators arranged on two-dimensional plane was systematically investigated by using the surface acoustic wave microfluidic technology. A robust synchronization of the oscillators was found at an input proper radio frequency (RF) power. The frequency of the chemical waves was increased with the increasing input power



*Dameng Guo, Yongqing Fu, Bo Zheng\**

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**Title**

Synchronization of Coupled  
Oscillators on a Two-  
Dimensional Plane